# Nonlinear optics with less than one photon

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## Abstract

We demonstrate suppression and enhancement of spontaneous parametric down-conversion via quantum interference with two weak fields from a local oscillator (LO). Pairs of LO photons are observed to upconvert with high efficiency for appropriate phase settings, exhibiting an effective nonlinearity enhanced by at least 10 orders of magnitude. This constitutes a two-photon switch, and promises to be useful for a variety of nonlinear optical effects at the quantum level.

Nonlinear effects in optics are typically limited to the high-intensity regime, due to the weak nonlinear response of even the best materials. An important exception occurs for resonantly enhanced nonlinearities, but these are restricted to narrow bandwidths. Nonlinear effects which are significant in the low-photon-number regime would open the door to a field of *quantum* nonlinear optics. This could lead to optical switches effective at the two-photon level (i.e. all-optical quantum logic gates); quantum solitons (e.g. two-photon bound states [1]); and a host of other phenomena. In this experiment, we demonstrate an effective two-photon nonlinearity mediated by a strong classical field. Quantum logic operations have already been performed in certain systems including trapped ions [2], NMR [3], and cavity QED [4], but there may be advantages to performing such operations in an all-optical scheme – including scalability and relatively low decoherence. A few schemes have been proposed for producing the enormous nonlinear optical responses necessary to perform quantum logic at the single-photon level. Such schemes involve coherent atomic effects (slow light [5] and E.I.T. [6]) or photon-exchange interactions [7]. We recently demonstrated that photodetection exhibits a strong two-photon nonlinearity [8], but this is not a coherent response, as it is connected to the amplification stage of measurement. While there has been considerable progress in these areas, coherent nonlinear optical effects have not yet been observed at the single-photon level. In a typical setup for second-harmonic generation, for instance a peak intensity on the order of 1 GW/cm$^2$ is required to provide an upconversion efficiency on the order of 10%. In the experiment we describe here, beams with peak intensities on the order of 1 mW/cm$^2$ undergo second-harmonic generation with an efficiency of about 1% – roughly 11 orders of magnitude higher than would be expected without any enhancement.

Our experiment relies on the process of spontaneous parametric down-conversion. If a strong laser beam with a frequency $2\omega$ passes through a material with a nonzero second-order susceptibility, $\chi^{(2)}$, then pairs of photons with nearly degenerate frequencies, $\omega$, can be created. In past experiments, interference phenomena have been observed between weak classical beams and pairs of down-conversion beams [9,?,?]. Although spontaneously down-converted photons have no well-defined phase (and therefore do not display first-order interference), the *sum* of the phases of the two beams is fixed by the phase of the pump. Koashi et al. [10] have observed this phase relationship experimentally using a local oscillator (LO) harmonically related to the pump. More recently Kuzmich et al. [11] have performed homodyne measurements to directly demonstrate the anticorrelation of the down-converted beams' phases. Some proposals for tests of nonlocality [12] have relied on the same sort of effect. Such experiments involve beating the down-converted light against a local oscillator at one or more beam splitters, and hence have multiple output ports. The interference causes the photon-correlations to shift among the various output ports of the beam splitters.

In contrast, in this experiment the actual photon-pair production rate is modulated. A simplified cartoon schematic of our experiment is shown in Fig. 1. A nonlinear crystal is pumped by a strong classical field, creating pairs of down-converted photons in two distinct modes (solid lines). Local oscillator beams are superposed on top of the down-conversion modes through the nonlinear crystal and are shown as dashed lines. A single-photon counting module (SPCM) is placed in the path of each mode. To lowest order there are two Feynman paths that can lead to both detectors firing at the same time (a coincidence event). A coincidence count can occur either from a down-conversion event (Fig. 1b.), or from a pair of LO photons (Fig. 1c.). Interference occurs between these two possible

paths provided they are indistinguishable. Depending on the phase difference between these two paths ($\varphi$), we observe enhancement or suppression of the coincidence rate. A phase-dependent rate of photon-pair production has been observed in a previous experiment using two pairs of down-converted beams from the same crystal [13]. By contrast, our experiment uses two independent LO fields which can be from classical or quantum sources, and subject to external control. If the phase between the paths (Fig. 1b, 1c) is chosen such that coincidences are eliminated, then photon pairs are removed from the LO beams by upconversion into the pump mode. If, however, one of the LO beams is blocked, then those photons that would have been upconverted are now transmitted through the crystal. This constitutes an optical switch in which the presence of one LO field controls the transmission of the other LO field – even when there is less than one photon in the crystal at a time. This switch does have certain limitations. First, it is inherently noisy due to the spontaneous down-conversion, which leads to coincidences even if both LO beams are blocked. Second, since the switch relies on interference, and hence phase, it does not occur between photon pairs but between the *amplitudes* to have a photon pair. While this may limit the usefulness of the effect as the basis of a "photon-transistor," a simple extension should allow it to be used for conditional-phase operations.

In order for the down-conversion beams to interfere with the laser beams, they must be indistinguishable in all ways (including frequency, time, spatial mode, and polarization). Down-conversion is inherently broadband and exhibits strong temporal correlations; the LOs must therefore consist of broadband pulses as well. We use a modelocked Titanium:Sapphire (Ti:Sa) laser operating with a central wavelength of 810 nm (Fig. 2). It produces 50-fs pulses at a rate of 80 MHz. This produces the LO beams, and its second-harmonic serves as the pump for the downconversion. Thus the downconversion is centered at the same frequency as the LO, and the LOs and the downconverted beams have similar bandwidths of around 30 nm. To further improve the frequency overlap, we frequency post-select the beams using a narrow bandpass (10-nm) interference filter [14]. As this is narrower than the bandwidth of the pump, it erases any frequency correlations between the downconversion beams. This removes the frequency correlations between the down-conversion beams. In addition to spectral indistinguishability, the two light sources must possess spatial indistinguishability. The down-conversion beams contain strong spatial correlations between the correlated photon pairs; measurement of a photon in one beam yields some information about the photon in the other beam. Such information does not exist within a laser beam; since there is only a single transverse mode, the photons must effectively be in a product state and exhibit no correlations. We therefore select a single spatial mode of the down-converted light by employing a simple spatial filter. The beams are focused onto a 25-$\mu$m diameter circular pinhole. The light that passes through the pinhole and a 2-mm diameter iris placed 5 cm downstream is collimated using a 5-cm lens. In order to increase the flux of down-converted photons into this spatial mode, we used a pump focusing technique related to the one demonstrated by Monken et al. [15]. The pump laser was focused directly onto the down-conversion crystal. Since the coherence area of the down-converted beams is set by the phase-matching acceptance angle, the smaller pump area reduced the number of spatial modes being generated at the crystal, improving the efficiency of selection in a single mode. Imaging the small illuminated spot of our crystal onto the pinhole, we were able to improve the rate of coincidences after the spatial filter by a factor of 30.

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The final condition necessary to obtain interference is to have a well-defined phase relationship between the LO beams and the down-conversion beams. To achieve this, the same Ti:Sa source laser is split into two different paths (Fig. 2.). The majority of the laser power (90%) is transmitted through BS1 into path 1, where it is type-I frequency-doubled to produce the strong (approximately 10-mW) classical pump beam with a central frequency of 405 nm. This beam is used to pump our down-conversion crystal (DC) after the 810-nm fundamental light is removed by colored glass filters. Instead of using down-conversion with spatially separate modes as shown in Fig. 1, we use type-II down-conversion, in which the photon pairs are emitted in the same direction but with distinct polarizations. The photon pairs are subsequently spatially filtered, spectrally filtered, and then split up by the polarizing beam splitter (PBS). The horizontally-polarized photon is transmitted to detector A, and the vertically-polarized photon is reflected to detector B. Detectors A and B are both single-photon counting modules (EG&G models SPCM-AQ-131 and SPCM-AQR-13). Path 1 also contains a trombone delay arm which can be displaced to change the relative phase between paths 1 and 2. To create the LO laser beams, we use the 10% reflection from BS1 into path 2. The vertically-polarized laser light is attenuated to the single-photon level by a set of neutral-density (ND) filters, and its polarization is then rotated by 45° using a zero-order half-wave plate, so that it serves simultaneously as LO for the horizontal and vertical beams. After the wave-plate, the light passes through a polarizer, which can be used to block one or both of the polarizations from this path. This is equivalent to blocking one or both of the LO beams. Ten percent of the light from path 2 is superposed with the down-conversion pump from path 1 at BS 2. The LO beams are thus subject to the same spatial and spectral filtering as the down-conversion and are separated by their polarizations at the PBS.

To demonstrate the interference effect, we adjusted the ND filters so that the coincidence rate from the down-conversion path was equal to the coincidence rate from the laser path. The polarizer was set to 45°, to transmit both horizontally and vertically polarized LOs. The singles rates from the down-conversion path alone were $770 \text{ s}^{-1}$ and $470 \text{ s}^{-1}$ for detectors A and B respectively, and the coincidence rate was $(3.3 \pm 0.3) \text{ s}^{-1}$ (the ambient background rates of roughly $6000 \text{ s}^{-1}$ for detector A and $8000 \text{ s}^{-1}$ for detector B have been subtracted from the singles rates, but no background subtraction is performed for the coincidences). The singles rates from the LO paths were $11800 \text{ s}^{-1}$ and $53800 \text{ s}^{-1}$ for detectors A and B respectively, and the coincidence rate from this path is $(3.3 \pm 0.3) \text{ s}^{-1}$. The LO intensities need to be much higher than the down-conversion intensities to achieve the same rate of coincidences because the photons in the LO beams are uncorrelated. As the trombone arm was moved to change the optical delay, we observed a modulation in the coincidence rate (Fig. 3.). The visibility of these fringes is $(48 \pm 1)\%$, and when we correct for our background ("accidental") coincidences the visibility is 57%. In theory, this visibility asymptotically approaches 100% in the very weak beam limit for equal coincidence rates from the down-conversion and the LO paths. At the peak of this fringe pattern, the total rate of photon pair production is greater than the sum of the rates from the independent paths. This is an enhancement of the rate of photon pair production. At the valley of the fringe pattern, the rate of the photon pair production is similarly suppressed. The spacing of the fringes is at the period for the 405-nm pump laser (approximately 1.3 fs/fringe).

The interference in the coincidence rate has been described as an enhancement or a

suppression in the rate of photon-pair production and because of this there should be an accompanying change in the *intensity* of the light reaching the detectors, and not merely the coincidence rate. Fig. 4. shows four sets of singles-rate data for detector A corresponding to four different polarizer settings. Recall that the light was incident upon the polarizer at 45°, so when the polarizer is set to 45°, both of the LO beams are free to pass. When the polarizer is set to 0° or 90°, one of the LO beams is blocked, and when the polarizer is set to -45° both of the LO beams are blocked. The left hand side of Fig. 4 shows the data for the two orthogonal diagonal settings of the polarizer, -45° (top) and 45° (bottom); the right hand side shows the data for the two orthogonal rectilinear settings, 0° (top) and 90° (bottom). For the 45° data, the singles rate at detector A shows fringes with a visibility of about 0.7%. This visibility is roughly 100 times smaller than the corresponding visibility in the coincidence rate because only about 1% of detected photons are members of a pair, due to the classical nature of our LO beams. The fringe spacing in the singles rate corresponds to that of the pump laser light at 405 nm even though it is the 810-nm intensity that is being monitored. By examining the other three polarizer settings (-45°, 0°, and 90°), it is apparent that in order to observe fringes in the singles rate, both LO paths must be open. This demonstrates that we are observing a nonlinear effect of one polarization mode on another, at the single-photon level.

When destructive interference reduces the intensity of the beams reaching the detectors, energy conservation dictates that all incident laser photon pairs must be undergoing sum frequenc generation. To explicitly verify that photon pairs are actually removed from the LO beams, a simple extension was performed. In the presence of the strong classical pump, it would be impossible to observe the upconverted photon directly, so we measure the reduction in the coincidence rate relative to the coincidence rate from the LOs alone. In order to maximize the effect, the coincidence rates from the LO path and the down-conversion path were set to $(38.2 \pm 0.7)$ s$^{-1}$ and $(1.2 \pm 0.2)$ s$^{-1}$, respectively. The coincidence rate was again recorded as a function of the optical delay and is shown by the filled circles in Fig. 5. A sinusoidal fit to the data is shown as a heavy black line, and has a fringe visibility of $(19.0 \pm 0.5)$ %. The coincidence rate from the LO paths alone was measured before and after the experiment was performed and is shown as a horizontal dashed line, as well as an open square with error bars indicated. For delay positions where the solid black line drops below the dashed line, the photon pair production rate drops below the rate from the LO rate alone. This reduction in the pairs is due to photon pairs being removed from the LO beams undergoing sum-frequency generation. From the fringe visibility, we can infer that at least $(15.7 \pm 1.7)$ % of the photon pairs from the local oscillator were converted into the second harmonic. This corresponds roughly to a few tenths of a percent of the photons overall.

We have demonstrated a quantum interference effect which is an effective nonlinearity at the single-photon level. We have shown that pairs of photons may be removed from a pair of LO beams, and that their removal results in the reduction of the number of photon pairs reaching a pair of detectors. This effect is accompanied by a corresponding change in the intensity of the beams. The effect studied in this work is closely analogous to second-harmonic generation in traditional nonlinear optical materials, but is enhanced by the simultaneous presence of a strong classical pump with appropriately chosen phase. For a different choice of phase, it should be possible to observe an effect analogous to cross-

phase modulation. Such a conditional phase shift may contribute to the realization of a controlled-phase gate for photons. These nonlinearities may also be useful for the problem of distinguishing the four maximally-entangled Bell-states that are ubiquitous in quantum optics. It has been shown that it is impossible to distinguish between all four states with only linear optics [16]. Work on unconditional teleportation [17] has been limited to efficiencies near $10^{-12}$. It may be possible to build a scheme centered around an effect like the one demonstrated in this work that would be capable of distinguishing all four states. Overall, effects such as this hold great promise for extending the field of nonlinear optics into the quantum domain.

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Fig. 1. A simplified cartoon of our experiment. a) Pairs of weak coherent states, or local oscillator (LO) beams (shown by dashed lines) are overlapped with the pair of down-converted photon beams. A coincidence count is registered either if b) a down-conversion event occurs, *or* if c) a pair of laser photons reaches the detectors (SPCMs). The $e^{i\varphi}$ in figure c) represents a controllable relative phase between the two Feynman paths that lead to a coincidence.

Fig. 2. A schematic for the setup of the experiment. BS 1 and BS 2 are 90/10 (T/R) beamsplitters; SHG consists of 2 lenses and a BBO nonlinear crystal for type-I second

harmonic generation; BG 39 is a coloured glass filter; ND is a set of neutral density filters; $\lambda/2$ is a zero-order half-wave plate; DC is a type-II down-conversion crystal; PH is a 25-$\mu$m diameter circular pinhole; I.F. is 10-nm-bandwidth interference filter, PBS is a polarizing beam splitter; and Det. A and Det. B are single-photon counting modules. The thinner solid line shows the beam path of the 810-nm light, and the heavier solid line shows the path of the 405-nm light.

Fig. 3. The coincidence rate as a function of the delay time. The interference is a phase-dependent enhancement or suppression of the photon pairs emitted from the crystal. The visibility of these fringes is $(47.4 \pm 0.9)\%$, and once corrected for background the visibility is 57%.

Fig. 4. The singles rate at detector A versus the delay for 4 different polarizer angle settings. The left-hand data sets are for the polarizer settings of $\pm 45°$. The right-hand column is for the polarizer settings of $0°$ and $90°$. The fringes are apparent only for the $+45°$ polarizer setting, and have a visibility of 0.7%. These four data sets show that both horizontally *and* vertically-polarized photons must be present for the effect to occur.

Fig. 5. The coincidence rate versus the delay for a case where the rates from the different Feynman paths are severely imbalanced. This demonstrates that some photon pairs from the LO beams are being upconverted. The average value of the coincidence rate from the laser path alone is represented by a hollow square and the dashed horizontal line. The solid curve is a sinusoidal fit to our data. It is apparent for certain delay (phase) settings the coincidence rate drops below the rate of coincidences from the LO paths alone. Based on the magnitude of this drop, we conclude that at least $(15.7 \pm 1.7)\%$ of the photon pairs from

the laser are upconverted.

a)

LO, ω

Pump, 2ω

$\chi^{(2)}$

ω

SPCM

SPCM

ω

LO, ω

b)

+

c)

$e^{i\varphi}$